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Sources of organochlorine contaminants and mercury in seabirds from the Aleutian archipelago of Alaska: Inferences from spatial and trophic variation

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ABSTRACT

Persistent organochlorine compounds and mercury (Hg) have been detected in numerous coastal organisms of the Aleutian archipelago of Alaska, yet sources of these contaminants are unclear. We collected glaucous-winged gulls, northern fulmars, and tufted puffins along a natural longitudinal gradient across the western and central Aleutian Islands (Buldir, Kiska, Amchitka, Adak), and an additional 8 seabird species representing different foraging and migratory guilds from Buldir Island to evaluate: 1) point source input from former military installations, 2) westward increases in contaminant concentrations suggestive of distant source input, and 3) effects of trophic status ($\delta^{15}N$) and carbon source ($\delta^{13}C$) on contaminant accumulation. Concentrations of Σ polychlorinated biphenyls (PCBs) and most chlorinated pesticides in glaucous-winged gulls consistently exhibited a 'U'-shaped pattern of high levels at Buldir and the east side of Adak and low levels at Kiska and Amchitka. In contrast, concentrations of Σ PCBs and chlorinated pesticides in northern fulmars and tufted puffins did not differ among islands. Hg concentrations increased westward in glaucous-winged gulls and were highest in northern fulmars from Buldir. Among species collected only at Buldir, Hg was notably elevated in pelagic cormorants, and relatively high Σ PCBs were detected in black-legged kittiwakes. Concentrations of Σ PCBs, dichlorodiphenyldichloroethylene (*p,p′* DDE), and Hg were positively correlated with $\delta^{15}N$ across all seabird species, indicating biomagnification across trophic levels. The east side of Adak Island (a former military installation) was a likely point source of Σ PCBs and *p,p′* DDE, particularly in glaucous-winged gulls. In contrast, elevated levels of these contaminants and Hg, along with PCB congener and chlorinated pesticide compositional patterns detected at Buldir Island indicated exposure from distant sources influenced by a combination of atmospheric–oceanic processes and the migratory movements of seabirds.

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1. Introduction

Elevated levels of persistent organic pollutants and heavy metals have been detected at high arctic latitudes such that these ecosystems are no longer considered pristine (AMAP, 1998). In particular, contaminants have been measured in a number of seabird species that inhabit the high arctic of Canada (e.g. Braune et al., 2005, Campbell et al., 2005) and Eurasia (e.g. Gabrielsen et al., 1995; Mehlum and Daelemans, 1995; Borga et al., 2001). Exposure to contaminants in arctic seabirds likely is derived mostly from atmospheric processes, oceanic currents, and riverine input which transport contaminants from industrialized latitudes (Barrie et al., 1992; Simonich and Hites, 1995). Migration to industrialized latitudes may also result in accumulation of contaminants in body burdens of seabirds, which ultimately may be deposited in arctic food webs upon their return (Blais, 2005; Blais et al., 2005). In turn, these contaminants are often biomagnified through the arctic food web (Hobson et al., 2002; Hoekstra et al., 2003).

Despite extensive published reports on contaminant levels in seabirds from the high arctic, comparatively little contaminant data exist for the diverse community of seabirds inhabiting mid-latitudinal regions of the North Pacific and Bering Sea, including the Aleutian archipelago. Biota inhabiting the Aleutians probably are exposed to contaminants of distant origin owing to the intensive atmospheric and oceanic processes (Stabeno et al., 1999) characteristic of the region as well as to contaminants emanating from point sources associated with military installations from World War II and the Cold War (AMAP, 1998; Anthony et al., 1999, 2007; Rocque and Winker, 2004). Furthermore, the archipelago supports 26 species comprised of over 10 million seabirds (Byrd et al., 2005) that play an integral role in nutrient cycling between oceanic and terrestrial systems (Croll et al., 2005). This diverse seabird community, which forages at multiple trophic levels and whose overwintering movements range from semi-resident to trans-equatorial migrants, is a valuable indicator of oceanic health (Mallory et al., 2006).

Previous studies indicated elevated levels of polychlorinated biphenyls (PCBs), dichlorodiphenyldichloroethylene (*p,p′* DDE), and mercury (Hg) in bald eagles (*Haliaeetus leucocephalus*) breeding in the central and western Aleutians (Anthony et al., 1999) and throughout the entire archipelago (Anthony et al., 2007). Point sources were implicated as the likely cause of high concentrations of PCBs in bald eagle eggs near former military installations on Adak and Amchitka islands. Elevated concentrations of PCBs also were reported in sea otters (*Enhydra lutris*) from these islands (Bacon et al., 1999). Conversely, certain chlorinated pesticides (mainly *p,p′* DDE) and Hg were highest in bald eagles from the western Aleutians and steadily increased westward, which suggested non-point sources presumably associated with atmospheric and/or oceanic currents. Contaminant patterns in pelagic cormorants (*Phalacrocorax pelagicus*) from the archipelago have revealed a similar westward increase in *p,p′* DDE (Rocque and Winker, 2004). Similarly, feathers from

Table 1 – Generalized life and natural history characteristics of seabird species sampled in the Aleutian archipelago of Alaska, 2000–01

Species	Islands sampled [a]	Diet [b,c]	Foraging location [c]	Overwintering status	Additional references
Glaucous-winged gull	BU, KI, AM, AD	Omnivorous	Nearshore	Likely resident, unknown proportion may overwinter in far western Pacific Ocean	Verbeek (1993), Irons and Williams (2007)
Northern fulmar	BU, KI, AM, AD	Omnivorous–offal	Pelagic	Non-resident, overwintering movements highly variable	Hatch and Nettleship (1998)
Tufted puffin	BU, KI, AM, AD	Fish–plankton	pelagic	Non-resident, overwinters pelagically in north Pacific Ocean	Piatt and Kitaysky (2002)
Black-legged kittiwake	BU	Fish–plankton	Pelagic	Non-resident, overwinters pelagically in north and central Pacific Ocean	Baird (1994)
Pelagic cormorant	BU	Fish	Nearshore	Likely resident	Hobson (1997)
Common murre	BU	Fish–plankton	Pelagic	Likely resident	Ainley et al. (2002)
Short-tailed shearwater	BU	Plankton	Pelagic	Non-resident, migrates to southern hemisphere	Jahncke et al. (2005)
Pigeon guillemot	BU	Fish–plankton	Nearshore	Likely resident	Ewins (1993)
Crested auklet	BU	Plankton	Pelagic	Likely resident	Jones (1993)
Parakeet auklet	BU	Plankton	Nearshore	Non-resident, overwinters pelagically in north and central Pacific Ocean	Jones et al. (2001)
Whiskered auklet	BU	Plankton	Nearshore	Likely resident	Byrd and Williams (1993)
Leach's storm-petrel	BU	Plankton–fish–offal	Pelagic	Non-resident, overwinters pelagically as far south as the equator	Huntington et al. (1996)

[a] BU = Buldir, KI == Kiska, AM = Amchitka, AD = Adak.
[b] Listed in approximate order of predominance in diet.
[c] Adapted from Byrd et al. (2005) with exception of short-tailed shearwater.

pigeon guillemots (*Cepphus columba*) (Burger et al., 2007a) and muscle from glaucous-winged gulls (*Larus glaucescens*) from the western islands of Kiska and Amchitka contained elevated and perhaps risk-prone levels of Hg (Burger et al., 2007b). However, we know of no published studies that have quantified organochlorine compounds and Hg in a diverse array of seabird species across the central and western archipelago, which is necessary to further elucidate sources of these contaminants.

We quantified concentrations and composition of PCBs, organochlorine pesticides, and Hg in seabirds with different migratory patterns and trophic status from the central and western archipelago and tested the following predictions. First, we examined spatial variation in contaminant levels among species to partition point vs. distant sources. We predicted that seabirds from islands with former military activity (East Adak and Amchitka) would have higher contaminant concentrations, particularly persistent PCBs, which would indicate point sources. Conversely, a longitudinal gradient of higher contaminant levels in the more remote western islands (Buldir and Kiska) would indicate greater contributions from distant sources (Anthony et al., 1999, 2007; Rocque and Winker, 2004). We further partitioned point vs. distant sources of PCBs by examining patterns of PCB congener composition. Lower chlorinated PCB congeners are more easily volatilized and transported over long distances compared to persistent, highly chlorinated congeners (Van den Brink, 1997), although this pattern can be confounded by species-specific differences in PCB metabolism and biotransformation (Borga et al., 2005). Second, we examined whether stable isotope values of nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) in seabird tissues could explain additional variation in contaminant concentrations among seabirds. Sources of primary production for consumers in marine environments can be partitioned by $\delta^{13}C$ because nearshore benthic algae have more enriched $\delta^{13}C$ values than pelagic phytoplankton (France, 1995; Hobson et al., 2002). Nitrogen isotopes are an indicator of trophic status (Kelly, 2000), where a 3–4‰ increase equates to a full trophic level step in high latitude food webs (Hobson, 1993). We predicted contaminant concentrations would increase with $\delta^{15}N$, while higher contaminant concentrations associated with nearshore $\delta^{13}C$ signatures would indicate more localized exposure compared to pelagic signatures from more distant sources.

2. Methods

2.1. Sample collection

We collected adult glaucous-winged gulls, northern fulmars (*Fulmarus glacialis*), and tufted puffins (*Fratercula cirrhata*) from Buldir, Kiska, Amchitka, West Adak, and East Adak Islands in the western and central Aleutians from June–August, 2000–2001 (Table 1, Fig. 1). We divided Adak Island into 2 sides because the east side had a known point source of PCBs associated with a former Naval Air Station deactivated in 1997 (Anthony et al., 2007). All seabirds were collected using shotguns with non-toxic steel shot. To reduce analytical costs, we collected 2–3

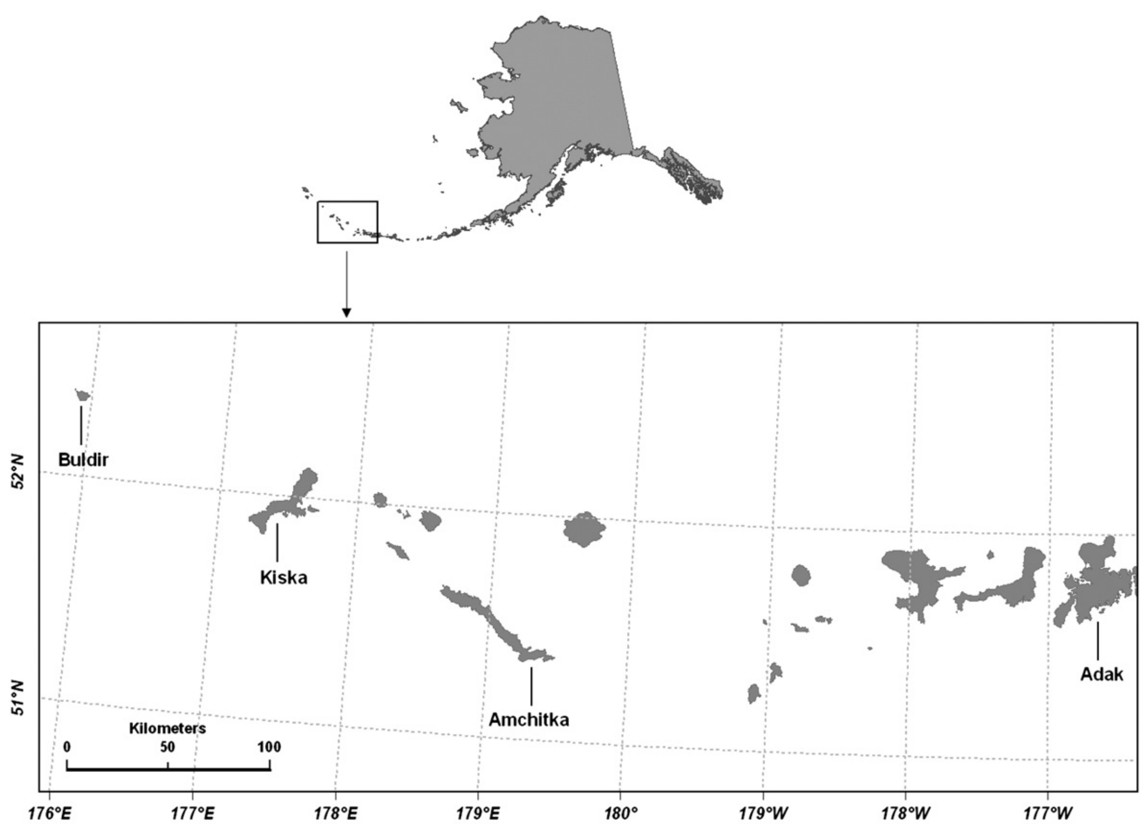

Fig. 1 – Map illustrating islands where seabird sampling occurred in the Aleutian archipelago of Alaska, 2000–01.

Table 2 – Geometric mean concentrations [a, b] (and range) of organochlorine compounds (µg/g, w.w.) and Hg (µg/g, d.w.) in composite liver samples from seabirds collected in the Aleutian archipelago of Alaska, 2000–01

Species	Island	*n*	% Lipid	Hg	Σ PCB	*p,p′* DDE	Σ DDT [c]	Σ CHLOR	Σ CBZ	Σ HCH	Σ DRIN	Σ OTH
Glaucous-winged Gull	Buldir	3	4.0 (3.3–4.6)	7.0 (6.8–7.3)	1.812 (1.511–2.357)	0.300 (0.169–0.437)	0.062 (0.047–0.094)	0.116 (0.109–0.133)	0.163 (0.111–0.334)	0.106 (0.077–0.145)	0.024 (0.021–0.030)	0.030 (0.020–0.042)
	Kiska	3	2.7 (2.2–3.0)	3.1 (1.8–4.5)	0.889 (0.750–1.032)	0.106 (0.079–0.182)	0.021 (0.012–0.048)	0.060 (0.056–0.065)	0.127 (0.083–0.160)	0.082 (0.054–0.104)	0.011 (0.007–0.032)	0.006 (0.004–0.011)
	Amchitka	3	2.8 (1.6–4.7)	1.4 (1.1–1.9)	0.569 (0.405–1.115)	0.061 (0.051–0.082)	0.002 (ND–0.003)	0.026 (0.023–0.028)	0.064 (0.061–0.072)	0.037 (0.036–0.042)	0.013 (0.010–0.017)	0.002 (0.002–0.004)
	West Adak	2	3.0 (2.9–3.1)	1.1 (1–1.1)	0.780 (0.673–0.906)	0.105 (0.071–0.155)	0.017 (0.010–0.029)	0.080 (0.073–0.090)	0.078 (0.063–0.098)	0.093 (0.074–0.117)	0.007 (0.007–0.008)	0.003 (ND–0.005)
	East Adak	2	5.0 (4.9–5.0)	1.0 (0.9–1.1)	2.452 (2.099–2.867)	0.342 (0.315–0.373)	0.007 (0.004–0.014)	0.131 (0.126–0.138)	0.217 (0.216–0.219)	0.142 (0.125–0.163)	0.013 (0.011–0.018)	0.014 (0.013–0.016)
	All Islands	13	3.4 (1.6–5.0)	2.2 (0.9–7.2)	1.083 (0.405–2.867)	0.142 (0.051–0.437)	0.015 (0.002–0.094)	0.068 (0.023–0.138)	0.116 (0.061–0.334)	0.080 (0.036–0.163)	0.013 (0.007–0.032)	0.008 (0.002–0.042)
Northern Fulmar	Buldir	3	3.0 (2.5–3.9)	24.6 (14.5–32.7)	0.698 (0.458–1.071)	0.134 (0.058–0.272)	0.018 (0.014–0.030)	0.119 (0.079–0.171)	0.036 (0.014–0.071)	0.036 (0.03–0.049)	0.010 (0.006–0.016)	0.01 (0.010–0.022)
	Kiska	2	2.3 (1.9–2.6)	10.1 (5.44–18.8)	0.782 (0.765–0.800)	0.144 (0.131–0.160)	0.017 (0.016–0.019)	0.099 (0.087–0.114)	0.068 (0.066–0.072)	0.074 (0.072–0.076)	ND (–)	0.011 (0.011–0.012)
	Amchitka	3	2.4 (2.1–2.7)	12.6 (6.0–21.0)	0.447 (0.362–0.507)	0.115 (0.104–0.130)	0.009 (0.007–0.013)	0.075 (0.063–0.086)	0.073 (0.067–0.084)	0.030 (0.022–0.038)	0.013 (0.01–0.016)	0.010 (0.009–0.012)
	West Adak	2	2.9 (2.4–3.4)	19.5 (15.5–24.6)	0.660 (0.561–0.777)	0.148 (0.148–0.149)	0.018 (0.012–0.029)	0.092 (0.091–0.094)	0.130 (0.095–0.179)	0.068 (0.068–0.068)	ND (–)	0.015 (0.012–0.02)
	East Adak	3	2.5 (2.3–2.8)	9.1 (6.8–11.5)	0.634 (0.598–0.681)	0.082 (0.080–0.087)	0.018 (0.016–0.020)	0.085 (0.080–0.089)	0.117 (0.100–0.155)	0.063 (0.057–0.070)	ND (–)	0.009 (0.009–0.010)
	All Islands	13	2.6 (1.9–3.9)	14.2 (5.4–32.7)	0.622 (0.362–1.070)	0.119 (0.058–0.272)	0.015 (0.007–0.030)	0.093 (0.063–0.171)	0.082 (0.044–0.179)	0.048 (0.022–0.076)	0.004 (0.005–0.016)	0.010 (0.003–0.022)
Tufted puffin	Buldir	3	3.4 (2.5–4.8)	2.4 (2.1–3.1)	0.319 (0.270–0.386)	0.039 (0.032–0.058)	0.012 (0.011–0.014)	0.032 (0.023–0.042)	0.044 (0.018–0.075)	0.024 (0.023–0.027)	0.006 (0.005–0.008)	0.002 (0.002–0.019)
	Kiska	3	2.0 (1.9–2.3)	3.1 (2.6–4.2)	0.244 (0.161–0.320)	0.030 (0.022–0.044)	0.004 (ND–0.007)	0.015 (0.011–0.020)	0.042 (0.033–0.052)	0.024 (0.018–0.030)	ND (–)	0.002 (ND–0.003)
	Amchitka	3	2.2 (1.6–3.3)	3.1 (2.7–3.3)	0.139 (0.083–0.299)	0.023 (0.019–0.036)	0.002 (ND–0.004)	0.006 (0.003–0.037)	0.025 (0.017–0.036)	0.012 (0.007–0.036)	0.002 (0.002–0.004)	0.002 (ND–0.006)
	West Adak	2	2.2 (1.9–2.4)	3.3 (2.5–4.4)	0.290 (0.228–0.371)	0.033 (0.028–0.040)	ND (–)	0.007 (ND–0.036)	0.079 (0.064–1.000)	0.032 (0.026–0.040)	0.009 (0.008–0.010)	ND (–)
	East Adak	2	2.7 (2.3–3.1)	2.9 (2.8–3.1)	0.552 (0.311–0.981)	0.041 (0.027–0.062)	0.008 (0.004–0.014)	0.027 (0007–0.104)	0.104 (0.061–0.177)	0.053 (0.022–0.128)	0.005 (ND–0.018)	0.002 (ND–0.006)
	All Islands	13	2.5 (1.6–4.8)	2.9 (2.1–4.4)	0.265 (0.083–0.981)	0.032 (0.019–0.062)	0.004 (0.003 –0.014)	0.017 (0.002–0.104)	0.048 (0.017–0.177)	0.024 (0.007–0.128)	0.004 (0.002–0.018)	0.002 (0.002–0.003111)

[a] For calculating geometric means, a value of 1/2 the LOD was substituted when no individual analyte comprising a particular organochlorine group was detected.% Lipid reported as arithmetic mean.
[b] ND — not detected.
[c] Not including *p,p′* DDE.

composite samples per species at each island. While composite sampling schemes can result in reduced statistical power due to smaller replicate sample sizes, values obtained from a composite sample represent an arithmetic average for the randomly collected individuals within the composite (Elder et al., 1980). Each composite sample comprised 3–5 individuals collected within distinct (ca. 1 km^2) areas separated from each other by >5 km. In order to further examine variation in contaminant burdens across a wider range of trophic levels and migratory guilds, one composite sample comprised of 4–5 individuals from each of the following adult species was collected at Buldir Island that supports the highest diversity of breeding seabirds in the Aleutians (Byrd et al., 2005): black-legged kittiwake (*Rissa tridactyla*), pelagic cormorant, Leach's storm-petrel (*Oceanodroma leucorhoa*), pigeon guillemot, common murre (*Uria aalge*), crested auklet (*Aethia cristatella*), whiskered auklet (*Aethia pygmaea*), parakeet auklet (*Aethia psittacula*), and short-tailed shearwater (*Puffinus tenuirostris*) (Table 1). Whole livers were excised from specimens using surgically clean scalpel blades, placed in certified chemically cleaned jars (VWR brand Trace Clean Quality Assured ®), and frozen at −20 °C until contaminant analysis. Variation in mass of individual livers comprising composite samples was low (mean CV=15%). We also collected a 6–7 g sample of pectoralis muscle from each bird for stable isotope analysis. Composite samples for stable isotopes matched those for contaminant samples for glaucous-winged gulls, northern fulmars, and tufted puffins at all islands. Isotope samples from seabirds collected only at Buldir were analyzed individually to further examine variation in trophic status and carbon sources, with the exception of whiskered auklets that were composited due to insufficient sample mass.

2.2. Analytical chemistry

Seabird liver samples were analyzed for 96 congener specific PCBs and 27 organochlorine compounds by the Geochemical and Environmental Research Group, Texas A&M University, College Station, Texas, USA. Total mercury (Hg) was analyzed by the Trace Element Research Laboratory, Texas A&M University, College Station, Texas, USA. Specific details regarding analytical chemistry are provided in Anthony et al. (2007). Quality control and assurance procedures and analytical results for both laboratories were approved by the U.S. Fish and Wildlife Service's Analytical Control Facility. Limits of detection averaged 0.96 ng/g for congeners (SD=0.3), 2.4 ng/g (SD=0.8) for organochlorine compounds, and 11.6 ng/g (SD=0.3) for Hg. Spiked recoveries for detected organic analytes averaged 86.8% (SD=21.7), and relative percent difference for duplicate samples averaged 14.0% (SD=14.7). For inorganic (Hg) analyses, spiked recoveries averaged 97.8% (SD=3.7), and relative percent difference averaged 3.7% (SD=3.4). Procedural blanks contained either no or insignificant traces of organic contaminants and Hg.

We analyzed stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) in pectoral muscle from the same individuals collected for contaminant analysis. A 6–7 g sample of muscle tissue was first dried in a 60 °C drying oven for 24–48 h, and then ground into a fine powder with a mortar and pestle. Lipids were extracted from all samples using methods described by

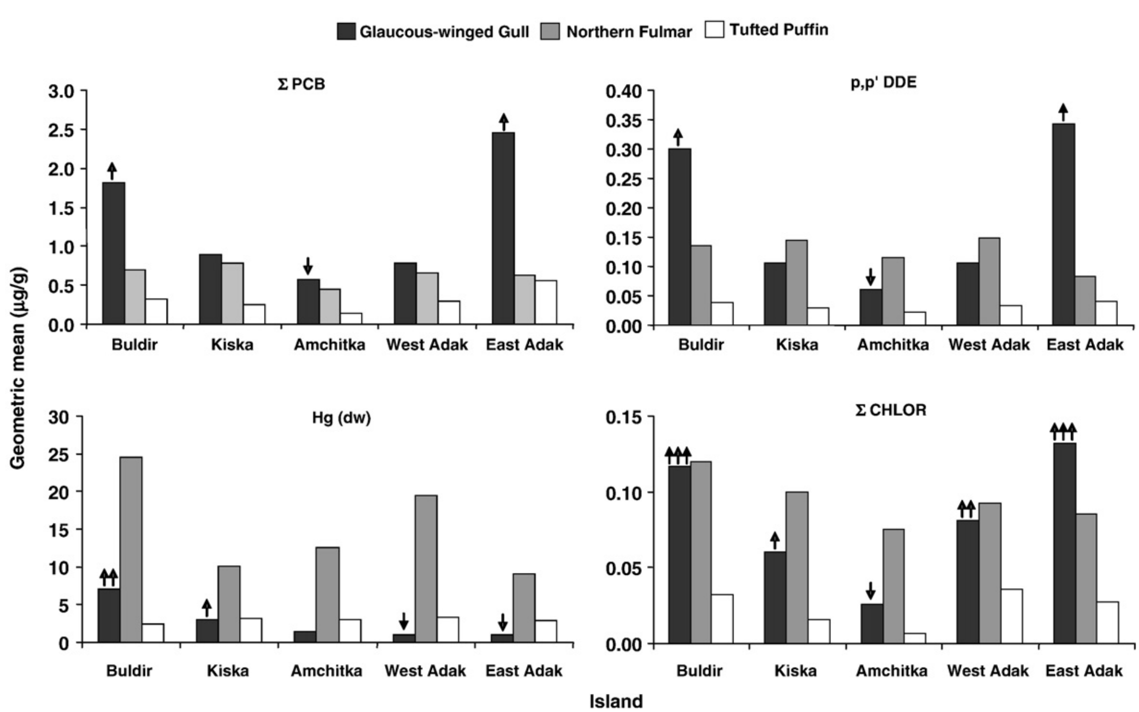

Fig. 2 – Hepatic concentrations (OCs = wet weight, Hg = dry weight) for primary contaminant groups in glaucous-winged gulls, northern fulmars, and tufted puffins collected in the Aleutian archipelago of Alaska, 2000–01. Arrows indicate how means differed significantly among islands for glaucous-winged gulls. Contaminant concentrations did not differ significantly among islands for northern fulmars or tufted puffins.

Dobush et al. (1985) to control for possible effects of high lipid content on carbon isotope ratios (Ricca et al., 2007). Lipid extraction did not affect $\delta^{15}N$ values (Ricca et al., 2007). Sample aliquots (1.0–1.5 mg) were sealed into 5×9 mm tin capsules and analyzed with a Europa Hydra 20/20 ® continuous flow isotope ratio mass spectrometer (Stable Isotope Facility, University of California, Davis, CA, USA). We expressed stable isotope ratios in standard delta notation:

$$\delta X = \left(\left(R_{\mathrm{sample}} / R_{\mathrm{standard}} \right) - 1 \right) \times 1000$$

where δX is the isotope ratio of the sample relative to the standard, and R_{sample} and R_{standard} are the fractions of the heavy to light isotopes in the sample and standard, respectively (i.e. $^{13}C/^{12}C$ and $^{15}N/^{14}N$) (Kelly, 2000). Values of δX are expressed in parts per thousands (‰). Measurement error determined from replicate analysis of internal laboratory standards was 0.19‰ for $\delta^{15}N$ and 0.04‰ for $\delta^{13}C$. Muscle isotopes from seabirds represent relatively short-term dietary integration (ca. 1 month) from the time of collection (Hobson, 1993).

2.3. Statistical analyses

For statistical hypothesis testing, we pooled concentrations of organochlorine compounds (OCs) to represent contaminant groups. Groups were Σ PCBs (equivalent to total aroclor content), *p,p′* DDE, Σ DDT (*p,p′* DDT, and DDD and *o,p′* DDE, DDT, and DDD), Σ chlordanes (CHLOR) (α and γ chlordane, oxychlordane, heptachlor, heptachlor epoxide, *cis* and *trans* nonachlor), Σ chlorobenzenes (CBZ) (hexachlorobenzene, 1,2,3,4 and 1,2,4,5-tetrachlorobenzene), Σ hexachlorocyclohexane (HCH) (α, β, δ, and γ HCH isomers), Σ drin pesticides (DRIN) (aldrin, dieldrin, endrin), Σ other compounds (OTH) (mirex, pentachloro-anisole, and endosulfan II), and Hg. For most statistical hypothesis testing, we separated *p,p′* DDE (hereafter DDE) from Σ DDT due to its well known ecological effects and environmental persistence, and for consistency with a related study of OCs in Aleutian bald eagles (Anthony et al., 2007). However, we included DDE in calculations of Σ DDT when describing percent composition of total organochlorine compounds, DDT isomers, and when testing for differences in ratios of DDE/Σ DDT. Only concentrations greater than the limit of detection (LOD) were included in calculations of group totals to avoid overestimation of rare compounds. We substituted a value of 1/2 the LOD for a single compound comprising an OC group when no compounds were detected. Non-detections were frequent only in the Σ DRIN and Σ OTH groups. We expressed OC concentrations on a wet weight basis (μg/g) and Hg on a dry weight basis (μg/g). Contaminant concentrations were $\log_e$ transformed prior to statistical analyses. Geometric means are reported for all concentrations unless noted otherwise.

We used a 2-factor MANOVA to test for overall differences in contaminant group concentrations among islands and species (glaucous-winged gulls, northern fulmars, and tufted puffins). When a significant interaction effect between islands and species was found, we analyzed differences among islands separated by species We used univariate ANOVA with Type III sums of squares to determine which contaminant groups differed significantly among islands when a significant difference was found among mean vectors, and then used Tukey–Kramer mean separation tests to determine how concentrations differed among islands. Because we were interested in detecting possible recent exposure to DDT in seabirds, we used 2-factor ANOVA with Type III sums of squares to compare arcsine-square root transformed DDE/Σ DDT ratios among islands and species. We used Principal Components Analysis (PCA) to characterize covarying combinations of 1) organochlorine contaminant groups and Hg, and 2) PCB congeners among species and islands. This analysis included seabird species only collected at Buldir and allowed more detailed examination of species patterns across all islands sampled. PCB congeners with a detection frequency of <50% as well as mono and di-chlorinated homologues were excluded from the congener PCA to minimize the influence of rarely detected congeners on the ordination.

We tested for differences in $\delta^{15}N$ and $\delta^{13}C$ values among species (glaucous-winged gulls, northern fulmars, and tufted puffins) and islands using a 2-factor ANOVA with Type III sums of squares. We regressed contaminant concentrations against $\delta^{15}N$ values using data pooled across all species, and used slope coefficients as estimates for biomagnification factors (BMFs) (e.g. Jarman et al., 1996; Borga et al., 2001;

Table 3 – Concentrations [a,b] of organochlorine compounds (μg/g, w.w.) and Hg (μg/g, d.w.) in composite liver samples from seabirds collected exclusively from Buldir Island in the Aleutian archipelago of Alaska, 2000–01

Species	n [c]	% Lipid	Hg	Σ PCB	*p,p′* DDE	Σ DDT [d]	Σ CHLOR	Σ CBZ	Σ HCH	Σ DRIN	Σ OTH
Black-legged kittiwake	1	4.1	3.340	0.890	0.022	0.028	0.060	0.063	0.050	0.010	0.009
Common murre	1	2.4	4.170	0.341	0.035	0.005	0.041	0.036	0.026	0.003	ND
Crested auklet	1	1.8	1.140	0.284	0.014	0.010	0.044	0.039	0.024	0.004	ND
Parakeet auklet	1	3.9	2.000	0.239	0.013	0.006	0.022	0.031	0.021	0.008	ND
Whiskered auklet	1	2.3	1.399	0.092	0.089	0.004	0.002	0.017	0.015	ND	ND
Pelagic cormorant	1	2.7	15.302	0.102	0.018	0.002	0.015	0.014	0.008	0.006	ND
Pigeon guillemot	1	3.8	6.360	0.150	0.026	ND	0.003	0.018	0.010	ND	ND
Short-tailed shearwater	1	2.3	0.729	0.201	0.006	0.008	0.026	0.045	0.016	0.004	0.002
Leach's storm-petrel	1	2.0	5.732	0.243	0.067	0.007	0.027	0.010	0.003	0.007	0.007

[a] For calculating geometric means, a value of 1/2 the LOD was substituted when no indivividual analyte comprising a particular organochlorine group was detected.% Lipid reported as arithmetic mean.
[b] ND — not detected.
[c] Composite sample comprised of 4–5 individuals.
[d] Not including *p,p′* DDE.

Buckman et al., 2004). If values of $\delta^{15}N$ and $\delta^{13}C$ were correlated, we then regressed $\delta^{13}C$ values against the residuals from the regressions of contaminant concentrations against $\delta^{15}N$ values to determine how the remaining variation not explained by trophic status was related to carbon source. We did not correct for island-specific variation in baseline isotopic signatures among islands (e.g. Jardine et al., 2006) because we lacked recent data for primary consumers. However, isotopic variation in several zooplankton species inhabiting the central and western Aleutians is low compared to isotopic variation across the entire archipelago and southern Bering Sea (Schell et al., 1998). Thus, we assumed differences in seabird $\delta^{15}N$ and $\delta^{13}C$ values among islands were not driven largely by local baseline isotopic variation.

3. Results

3.1. Organochlorine and Hg composition

Total PCBs comprised the largest proportion (62–69%) of total OCs in glaucous-winged gulls, northern fulmars, and tufted puffins at all islands, followed by Σ DDT (10–16%), Σ CBZ (5–12%), Σ CHLOR (3–11%), Σ HCH (1–8%), Σ DRIN (0–2%), and Σ OTH (0–1%). A similar pattern was detected in seabirds collected exclusively at Buldir, with the exception of whiskered auklets where Σ DDT (43%) and Σ PCBs (42%) comprised approximately equivalent proportions of Σ OCs. Mercury was detected in all samples. Within primary chlorinated pesticide groups (Σ DDT and Σ CHLOR), *p,p′* DDE comprised the majority (75–95%) of Σ DDT in glaucous-winged gulls, northern fulmars, and tufted puffins at all islands. Chlordane composition was more variable among species, where α-chlordane (48–68%) was the primary compound in glaucous-winged gulls and tufted puffins and oxychlordane (50–76%) the primary chlordane compound in northern fulmars. Gamma chlordane comprised only 3–13% of Σ CHLOR among these species. For species collected only at Buldir, *p,p′* DDE comprised the majority (89–100%) of Σ DDT in common murre, whiskered auklet, pelagic cormorant, pigeon guillemot, and Leach's storm-petrel, while *p,p′* DDD comprised a larger percentage of Σ DDT in short-tailed shearwater (47%) and black-legged kittiwake (39%). Chlordanes were comprised entirely of oxychlordane in pigeon guillemots and whiskered auklets, whereas α-chlordane (77–89%) was the dominant chlordane

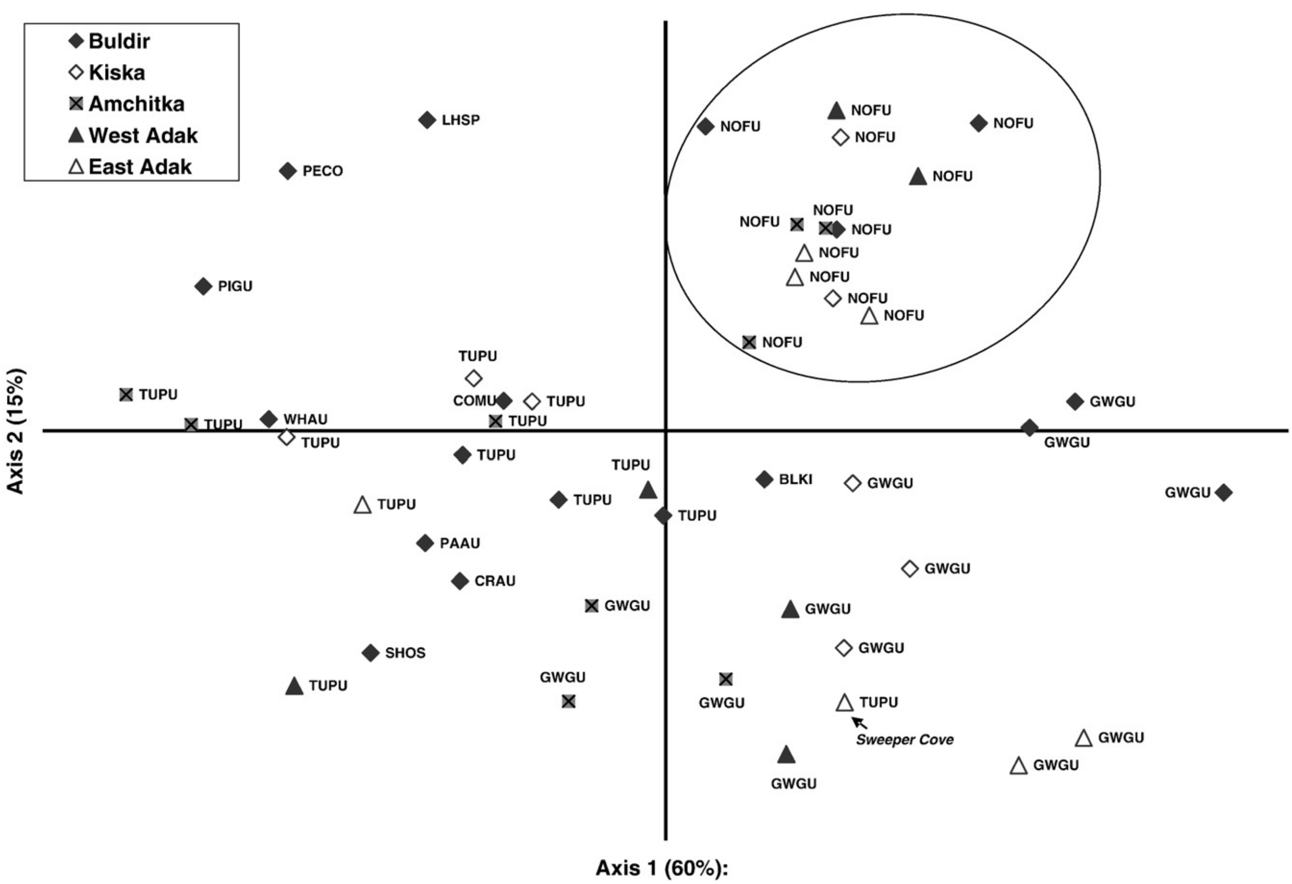

Fig. 3 – Principal components analysis for organochlorine compounds and Hg in all seabird liver samples collected (including those only collected at Buldir) in the Aleutian archipelago of Alaska, 2000–01. Axis 1 (horizontal axis) is positively associated with overall organochlorine contamination, and Axis 2 (vertical axis) is positively associated with Hg concentrations relative to OC contamination. Circle denotes grouping for northern fulmars. Species abbreviations are: glaucous-winged gull (GWGU), northern fulmar (NOFU), tufted puffin (TUPU), crested auklet (CRAU), parakeet auklet (PAAU), whiskered auklet (WHAU), black-legged kittiwake (BLKI), pelagic cormorant (PECO), pigeon guillemot (PIGU), short-tailed shearwater (SHOS), common murre (COMU), and Leach's storm-petrel (LHSP).

compound in parakeet auklets, crested auklet, short-tailed shearwaters, and common murres.

Significant variation in DDE/Σ DDT ratios was detected among species collected at all islands ($F_{[4,24]}=6.6$, $P=0.005$), but interpretation was confounded by a significant species*island interaction ($F_{[8,24]}=2.6$, $P=0.03$). Consequently, we conducted separate island comparisons for each species. Ratios of DDE/Σ DDT did not differ significantly among islands for glaucous-winged gulls ($\bar{x}_{range}=77$–96%, $F_{[4,8]}=2.2$, $P=0.15$), or northern fulmars ($\bar{x}_{range}=77$–88%, $F_{[4,\ 8]}=1.9$, $P=0.21$). Ratios of DDE/Σ DDT were marginally different among islands for tufted puffins ($F_{[4,8]}=3.7$, $P=0.06$) with suggestive evidence of lower proportions of DDE at Buldir ($\bar{x}$=68%) compared to other islands ($\bar{x}_{range}=76$–82%).

3.2. Organochlorine and Hg concentrations

Overall, contaminant group concentrations differed among species ($\lambda=0.02$, $P<0.0001$) and islands ($\lambda=0.009$, $P<0.0001$). However, differences in concentrations among islands were not consistent among species (island*species interaction: $\lambda=0.002$, $P<0.001$), so we tested for differences among islands separately by species. Percent lipid had no overall effect on contaminant group concentrations ($\lambda=0.49$, $P=0.17$) and was excluded from subsequent models.

Among contaminant groups comprising the majority of OC composition (hereafter primary contaminant groups) (Table 2), Σ PCBs, p,p' DDE, and Σ CHLOR in glaucous-winged gulls exhibited a significant 'U' shaped pattern ($F_{[4,8]}>7.9$, $P<0.007$) where concentrations were highest at Buldir (western-most island) and East Adak (eastern-most island) and lowest at Amchitka (Fig. 2). In contrast, Hg in glaucous-winged gulls increased significantly ($F_{[4,8]}=22.7$, $P=0.0002$) in a westerly direction (Fig. 2), yet concentrations of all primary contaminant groups did not differ among islands for northern fulmars ($F_{[4,8]}<1.8$, $P>0.22$) or tufted puffins ($F_{[4,8]}<2.4$, $P>0.13$). A notable yet statistically insignificant trend of high concentrations of Hg in northern fulmars occurred at Buldir and West Adak. Furthermore, average concentrations of Hg in northern fulmars across all islands were 5 times higher than glaucous-winged gulls and 6 times higher than tufted puffins. The only lesser contaminant groups in glaucous-winged gulls that differed among islands were Σ CBZ, Σ HCH, and Σ OTH ($F_{[4,8]}>4.3$, $P<0.04$) where concentrations were generally higher at East Adak and Buldir compared to Amchitka. For northern fulmars, Σ HCH concentrations were highest at Kiska compared to Amchitka ($F_{[4,8]}=10.5$, $P=0.003$) whereas Σ DRIN concentrations were highest at Amchitka and Buldir compared to Adak (both sides) and Kiska ($F_{[4,8]}=15.3$, $P=0.0008$). Lesser contaminant groups did not differ among islands in

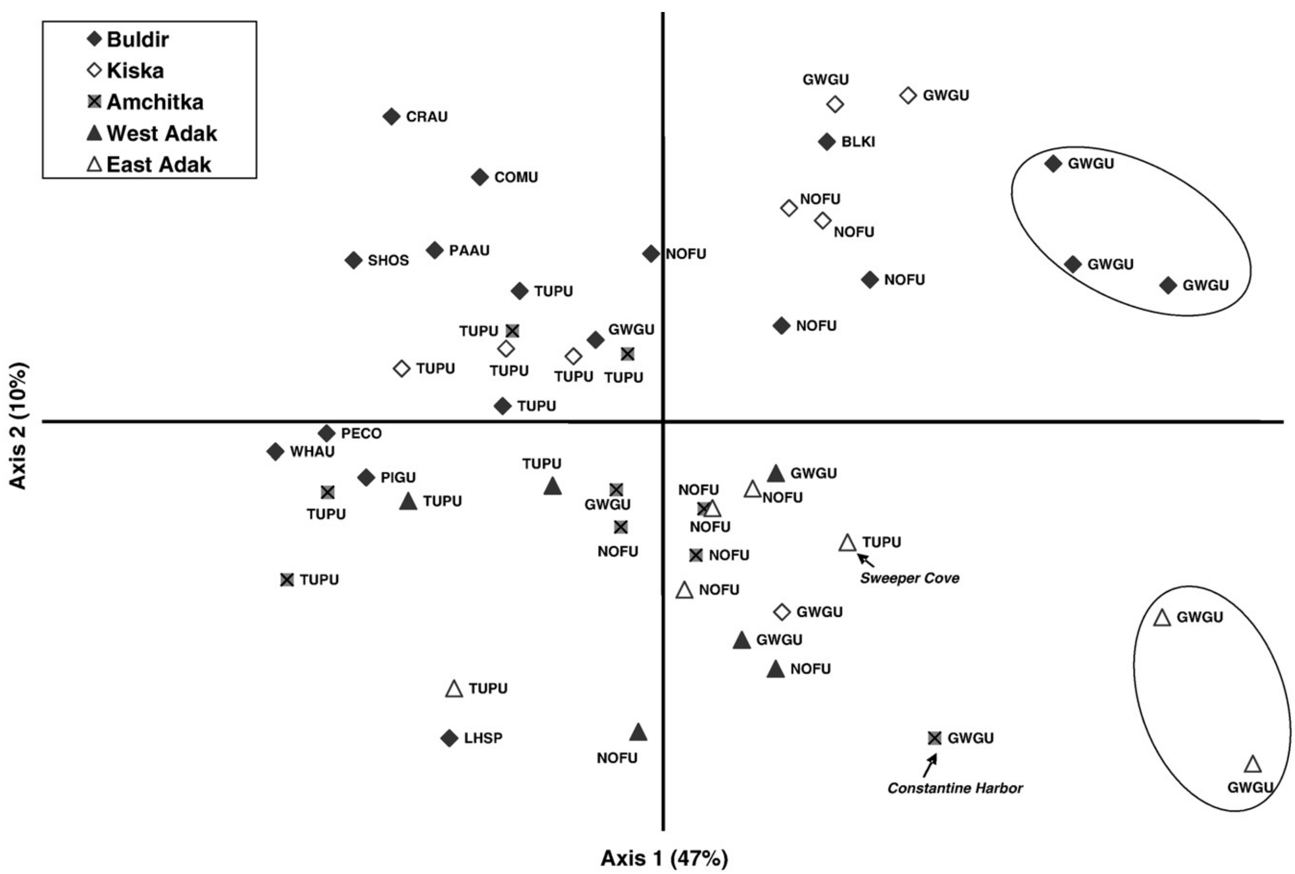

Fig. 4 – Principal components analysis for PCB congeners in all seabird liver samples collected (including those only collected at Buldir) from the Aleutian archipelago of Alaska, 2000–01. Axis 1 (horizontal axis) is positively associated with overall PCB contamination, and Axis 2 (vertical axis) is positively associated with high concentrations of lower chlorinated PCB congeners. Circles denote groupings for glaucous-winged gulls from East Adak and Buldir. Corresponding meanings for species abbreviations are provided in Fig. 3.

tufted puffins ($F_{[4,8]} \leq 3.5$, $P \geq 0.06$) (Table 2). Among seabirds collected only at Buldir, Σ PCBs were twice as high in black-legged kittiwakes compared to all other species (Table 3). Whiskered auklets and Leach's storm-petrel had the most elevated levels of *p,p′* DDE compared to all other species, while pelagic cormorants had notably elevated Hg followed by pigeon guillemots and Leach's storm-petrel. Concentrations of all other contaminants were relatively low in all species.

Island and species patterns were further illustrated by PCA, where 75% of the cumulative variance in OC group and Hg concentrations among species and islands was explained by the first 2 axes (Fig. 3). Axis 1 was positively correlated with increasing concentrations of all OCs, particularly Σ PCBs, *p,p′* DDE, and Σ CHLOR ($r \geq 0.85$). Axis 2 was positively correlated with Hg ($r = 0.88$) and negatively correlated with all OCs ($r \leq -0.38$). Therefore, Axis 1 represented a general measure of overall OC contamination, and Axis 2 indicated high Hg concentrations relative to OCs. Samples separated more distinctly by species than by island in the ordination space, although species from East Adak and glaucous-winged gulls from Buldir were mostly associated with higher overall OC concentrations. Glaucous-winged gulls and northern fulmars were associated with higher concentrations of OCs along Axis 1, and northern fulmars were associated most strongly with high Hg concentrations along Axis 2. Tufted puffins were generally associated with lower OC concentrations with the exception of one sample from East Adak collected near Sweeper Cove, a known site of PCB contamination (Anthony et al., 2007). Most species collected exclusively from Buldir were associated with lower OC concentrations, with the exception of black-legged kittiwakes. Notably, pelagic cormorants, Leach's storm-petrel, and pigeon guillemots were associated with high Hg and low OC concentrations.

3.3. PCB congeners

PCB congeners known to bioaccumulate in marine environments (IUPAC#s 101, 138, 153, 180, and 183) generally comprised a higher percentage ($\bar{x} = 37\%$, range: 25–50%) of Σ PCB congeners among seabirds. However, lighter and less chlorinated congeners (IUPAC#s 22, 28, 45, 47, 66, 69, 83, and 99) also comprised substantial percentages ($\bar{x} = 26\%$, range: 6–42%) of Σ PCB congeners.

The two principal components for PCB congeners among all seabird samples explained 57% of the cumulative variation in congener concentrations (Fig. 4). Axis 1 was positively correlated with increasing concentrations of all PCB congeners, particularly IUPAC#s 74, 99, 118, 138, 153, 167, 180, 194, 196, and 206 ($r \geq 0.75$). Axis 2 was positively correlated with lower chlorinated congeners such as IUPAC#s 26, 28, 47, 49, 69, and 83 ($r \geq 0.33$) and inversely related to IUPAC#s 105, 110, 138, 153, 170, 171, 180, and 191 ($r \leq -0.33$). Therefore, Axis 1 represented a general measure of overall PCB contamination, and Axis 2 represented a gradient of congener chlorination.

Table 4 – Descriptive statistics for carbon and nitrogen stable isotopes [a] from lipid extracted breast muscle samples from seabirds collected in the Aleutian archipelago of Alaska, 2000–01

Species	Island	n	δ^{15}N				δ^{13}C			
			Mean	SD	Min	Max	Mean	SD	Min	Max
Glaucous-winged gull	Buldir	3	12.0	0.2	11.9	12.2	-19.7^{b}	0.3	−19.9	−19.3
	Kiska	3	11.4	0.4	11.0	11.9	-18.8^{ab}	1.0	−19.9	−17.9
	Amchitka	3	11.4	0.7	10.5	12.0	-17.2^{a}	1.2	−18.3	−15.9
	West Adak	2	12.2	0.1	12.2	12.3	-18.8^{ab}	0.1	−18.8	−18.7
	East Adak	2	12.3	0.1	12.3	12.4	-18.4^{ab}	0.1	−18.5	−18.4
Northern fulmar	Buldir	3	13.3^{ab}	0.3	12.9	13.5	-20.4^{b}	0.1	−20.4	−20.3
	Kiska	2	13.1^{b}	0.0	13.1	13.1	-19.9^{a}	0.2	−20.0	−19.8
	Amchitka	3	12.8^{b}	0.3	12.4	13.0	-20.3^{b}	0.0	−20.3	−20.2
	West Adak	2	14.2^{a}	0.1	14.2	14.3	-20.1^{a}	0.3	−20.3	−19.9
	East Adak	3	12.9^{b}	0.4	12.6	13.4	-19.8^{a}	0.1	−19.9	−19.7
Tufted puffin	Buldir	3	10.6^{b}	0.1	10.5	10.8	-21.4^{b}	0.2	−21.6	−21.3
	Kiska	3	10.4^{b}	0.1	10.3	10.5	-21.8^{b}	0.1	−21.9	−21.7
	Amchitka	3	10.9^{b}	0.2	10.7	11.0	-20.8^{ab}	0.2	−21.0	−20.7
	West Adak	2	11.7^{a}	0.3	11.4	11.9	-20.0^{a}	0.7	−20.5	−19.5
	East Adak	2	11.5^{ab}	0.1	11.7	11.3	-19.8^{a}	0.8	−20.4	−19.2
Black-legged kittiwake	Buldir	4	11.3	0.1	11.1	11.5	−20.3	0.1	−20.4	−20.2
Common murre	Buldir	4	10.5	0.8	9.4	11.1	−20.6	0.2	−20.9	−20.4
Pelagic cormorant	Buldir	4	11.9	0.5	11.3	12.3	−18.6	1.7	−20.1	−16.8
Pigeon guillemot	Buldir	4	10.8	0.6	10.1	11.6	−17.8	0.6	−18.3	−17.1
Short-tailed shearwater	Buldir	5	9.3	0.5	8.8	9.9	−21.0	0.6	−21.7	−20.2
Crested auklet	Buldir	5	9.6	1.4	8.7	12.1	−22.4	0.7	−22.9	−21.2
Parakeet auklet	Buldir	4	9.8	0.1	9.7	9.8	−21.6	0.7	−22.5	−20.9
Leach's storm-petrel	Buldir	4	11.7	0.8	10.8	12.7	−20.8	0.2	−20.9	−20.5
Whiskered auklet	Buldir	1	9.8				−21.1			

Islands are ordered from west to east.
Values sharing the same letter for a particular species did not differ significantly.
[a] Values from species collected exclusively from Buldir, except Whiskered Auklets, are from individual samples. All other values represent composite samples.

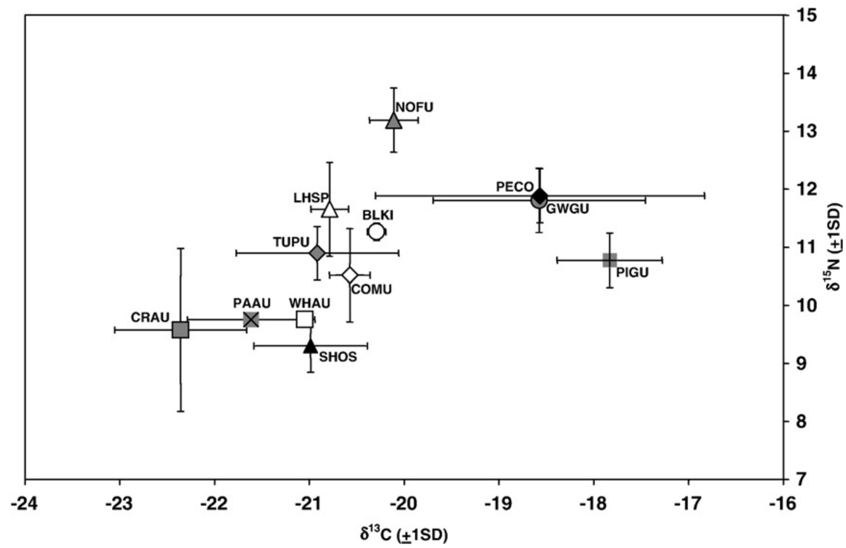

Fig. 5 – Mean values (±1 SD) for stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotopes in seabird muscle samples collected from the Aleutian archipelago of Alaska, 2000–01. Values for glaucous-winged gulls, northern fulmars, and tufted puffins are averaged across all islands. Corresponding meanings for species abbreviations are provided in Fig. 3.

Most glaucous-winged gulls, northern fulmars, and black-legged kittiwakes positively aligned with higher values along Axis 1 and did not overlap with most tufted puffins or any other seabirds collected only at Buldir. Differences in PCB congener concentrations among islands were demonstrated by the distinct separations along Axis 2 that indicated seabirds

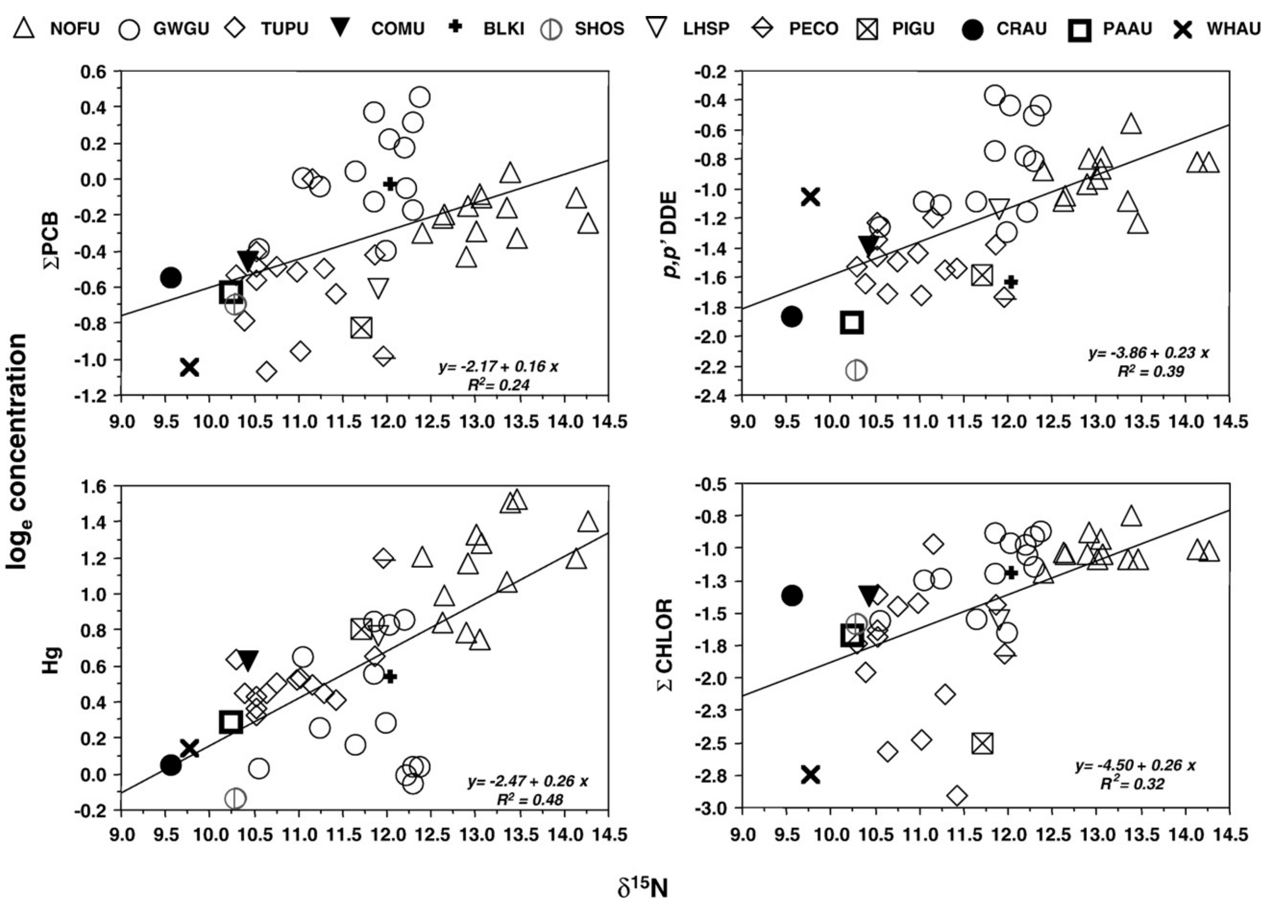

Fig. 6 – Relationships between concentrations of primary contaminant groups and stable nitrogen ($\delta^{15}N$) isotope values in seabirds sampled in the Aleutian archipelago of Alaska, 2000–01. Corresponding meanings for species abbreviations are provided in Fig. 3.

from Buldir (with the exception of Leach's storm-petrel) and Kiska were associated with higher concentrations of lower chlorinated PCB congeners than seabirds from Amchitka and both sides of Adak. Notably, glaucous-winged gulls from East Adak and Buldir were similar along Axis 1 but clearly separated along Axis 2. Glaucous-winged gulls collected from Amchitka's Constantine Harbor also were associated with higher concentrations of highly chlorinated PCB congeners than those collected at the 2 other Amchitka sites.

3.4. *Stable isotopes and relationships to contaminants*

There were significant island*species interactions for both $\delta^{15}N$ and $\delta^{13}C$ ($F_{[8,24]} \geq 2.6$, $P \leq 0.04$), so inter-island comparisons were conducted separately for each species (Table 4). Values of $\delta^{15}N$ did not differ significantly among islands for glaucous-winged gulls ($F_{[4,8]} = 2.8$, $P = 0.1$). However, values of $\delta^{15}N$ were most enriched in northern fulmars from West Adak, Kiska, and Buldir ($F_{[4,8]} = 7.7$, $P \geq 0.007$) and in tufted puffins from West Adak ($F_{[4,8]} = 18.8$, $P = 0.0004$). Values of $\delta^{13}C$ differed significantly among islands for all species ($F_{[4,8]} \geq 3.8$, $P \leq 0.05$). Values of $\delta^{13}C$ were enriched in glaucous-winged gulls at Amchitka compared to Buldir. Conversely, values of $\delta^{13}C$ were enriched in northern fulmars at West Adak, East Adak, and Kiska compared to Amchitka and Buldir, and in tufted puffins at West Adak compared to Amchitka, Buldir, and Kiska. These results indicated that island differences in stable isotope values did not occur incrementally with longitude with the exception of tufted puffins.

Stable isotope signatures indicated that northern fulmars foraged at the highest trophic position of any of the species studied and at locations intermediate between highly pelagic and nearshore (Fig. 5). Pelagic cormorants, pigeon guillemots, Leach's storm-petrel, and black-legged kittiwakes had $\delta^{15}N$ values similar to glaucous-winged gulls, while parakeet, crested, and whiskered auklets and short-tailed shearwaters foraged at slightly lower trophic positions than tufted puffins. Crested auklets had the most pelagic $\delta^{13}C$ signature (−23‰) of any seabird, while parakeet auklets, whiskered auklet, short-tailed shearwaters, and black-legged kittiwakes also had depleted $\delta^{13}C$ signatures similar to tufted puffins (−20 to −21‰), which indicated more pelagic feeding for these species. In contrast, glaucous-winged gulls, pelagic cormorants and pigeon guillemots had similar $\delta^{13}C$ values (−17 to −18) that signified more nearshore or benthic feeding as expected based on known life history (Table 1).

Values of $\delta^{15}N$ and $\delta^{13}C$ were positively correlated across all seabirds combined ($F_{[1,46]} = 6.8$, $P = 0.01$, $r^2 = 0.11$). Concentrations of primary contaminant groups and Hg were positively correlated with values of $\delta^{15}N$ (Fig. 6), which indicated varying degrees of biomagnification of these contaminants across

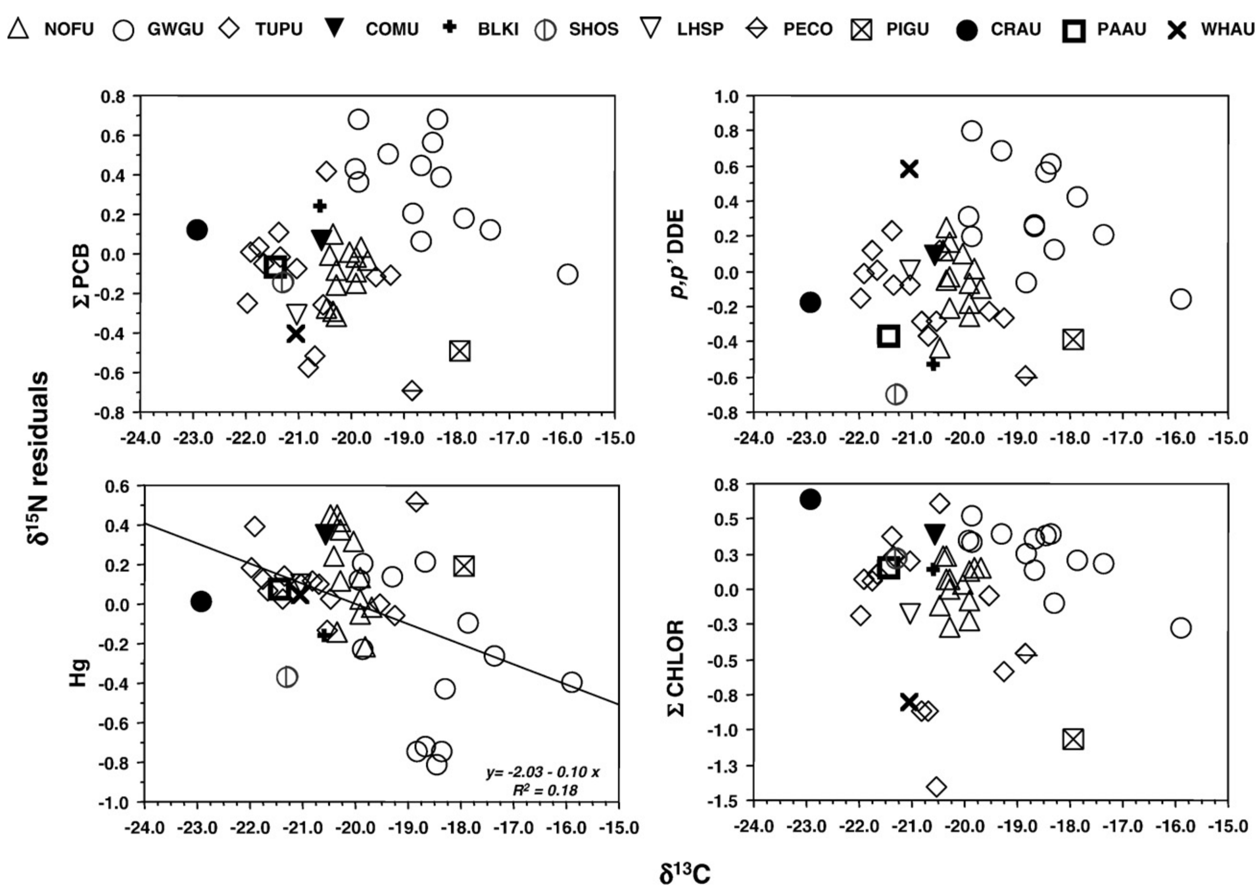

Fig. 7 – Relationships between residual variation unexplained by $\delta^{15}N$ in concentrations of primary contaminant groups and stable carbon ($\delta^{13}C$) isotope values in seabirds sampled in the Aleutian archipelago of Alaska, 2000–01. Corresponding meanings for species abbreviations are provided in Fig. 3.

seabird trophic levels. The relationship between Hg and δ^{15}N was the most predictable ($r^2=0.48$) and had the highest biomagnification factor (slope=0.26). Biomagnification factors for *p,p′* DDE and Σ CHLOR were similar to Hg but less predictable ($r^2<0.38$). Total PCBs had the weakest BMF (slope=0.16). The residual variation from the contaminant — δ^{15}N regressions was unrelated to values δ^{13}C for Σ PCBs, DDE, and Σ CHLOR ($F_{[1,46]}\leq 2.5$, $P>0.12$, $r^2\leq 0.05$) (Fig. 7). However, values of δ^{13}C were negatively correlated with Hg residuals ($F_{[1,46]}=10.3$, $P=0.002$, $r^2=018$), which indicated higher Hg for species foraging in pelagic compared to nearshore habitats after accounting for δ^{15}N (Fig. 7).

4. Discussion

Our results demonstrated that sources of environmental contaminants in seabirds inhabiting the Aleutian archipelago were highly complex in that they varied among species and islands, and both point and distant sources contributed to seabird contaminant exposure. However, notable patterns emerged.

4.1. *Geographical patterns*

As we predicted, point sources of contaminants were evident at East Adak, where concentrations of Σ PCBs and chlorinated pesticides such as *p,p′* DDE, Σ DDT, Σ CHLOR were elevated in glaucous-winged gulls in comparison to West Adak, Amchitka, and Kiska. The strength of East Adak as a point source, particularly for Σ PCBs, was supported by concentrations approximately 3 times lower in glaucous-winged gulls from the west side of Adak. Concentrations of Σ PCBs in glaucous-winged gulls from East Adak were similar to point source patterns detected in bald eagles (Anthony et al., 1999, 2007), blue mussels (S. Reese, M.S. thesis, University of California — Santa Cruz), fish (A. K. Miles, unpublished data), and pelagic cormorants (Rocque and Winker 2004) from East Adak. Also, the highest overall contaminant concentrations in tufted puffins were associated with the former military harbor (Sweeper Cove) on East Adak. In contrast, concentrations of Σ PCBs in glaucous-winged gulls were lowest at Amchitka, and the lowest mean concentrations of OCs in northern fulmars and tufted puffins, while not statistically different among islands, were also from Amchitka. We expected seabirds from Amchitka to have high concentrations of Σ PCBs owing to its recent military history as a site of underground nuclear testing during the 1970s. The comparably low OC concentrations detected in seabirds from Amchitka was surprising because bald eagles (Anthony et al., 1999, 2007), blue mussels (S. Reese, M.S. thesis, University of California — Santa Cruz,), and some fish species (A. K. Miles, unpublished data) from Amchitka had high concentrations of Σ PCBs and chlorinated pesticides similar to East Adak. Nevertheless, glaucous-winged gulls collected from Constantine Harbor (a heavily used harbor during the 1970s) had the highest PCB levels among all gull samples collected at Amchitka. Furthermore, a strong point source was not evident at Kiska, which had extensive military activity during WWII but has been largely unoccupied since that time. These contaminant patterns at Adak, Amchitka, and Kiska suggest that point source effects on seabirds appear highly localized and may decline in strength over time.

Alternatively, elevated levels of all primary contaminant groups in seabirds from Buldir may indicate input from distant sources. In particular, concentrations of Σ PCBs, *p,p′* DDE, and Σ CHLOR in glaucous-winged gulls were statistically similar to those detected in gulls from East Adak, yet Buldir is highly isolated with no history of military or industrial activity. Furthermore, mean concentrations of these contaminants declined in an easterly direction as predicted by our distant source hypothesis before increasing at East Adak. Long range transport via atmospheric and oceanic currents may provide further insight into this U shaped pattern. During winter, the Aleutian low pressure system pulls air from Eurasia across the Pacific Ocean and into the Bering Sea that is a probable source for volatile globally distilled contaminants (Barrie et al., 1992; AMAP, 1998). The role of ocean currents as a distant source from Eurasia is more complicated because the Alaska Stream flows westward along the Pacific side of the Aleutians and then enters the eastward-flowing North Slope Alaska Current along the Bering side primarily through Amukta Pass in the east and Amchitka Pass in the west (Stabeno et al., 1999). Movements by seabirds themselves may be another vector of contaminant transport (Blais, 2005). Actual proportions of Aleutian seabirds that may overwinter in Pacific Rim regions of point source contamination may be higher than is actually known (e.g. Irons and Williams, 2007). If true, then migratory movements may result in exposure to less volatile compounds that are not readily globally distilled such as *p,p′* DDE (Simonich and Hites, 1995), which would further explain elevated contaminant levels in seabirds from Buldir.

Mercury in glaucous-winged gulls was the only contaminant that increased westwardly and tracked patterns detected in bald eagles from the same islands (Anthony et al., 1999, 2007). High concentrations of Hg also were detected in northern fulmars, pelagic cormorants, and Leach's storm-petrel from Buldir. In contrast, this longitudinal pattern was not previously detected in pelagic cormorants from the archipelago that included islands sampled further to the east and west compared to our study (Rocque and Winker, 2004), or in glaucous-winged gull and tufted puffins from Kiska, Amchitka, and Adak (Burger et al., 2007b). A key difference between studies is that our data set included samples from Buldir; an island where Hg concentrations in seabirds have not been reported previously. However, Rocque and Winker (2004) did detect high concentrations of Hg in pelagic cormorants from Attu, which is the western-most island in the archipelago (ca. 175 km west of Buldir), and also detected westward increases in concentrations of other trace elements. Their study along with our results lends further support for high Hg concentrations in seabirds from western islands. Naturally derived inorganic Hg from cinnabar and volcanic eruptions along the Aleutians likely provides some local input (AMAP 1998; Rocque and Winker, 2004) but is an unlikely explanation for higher levels of Hg at the western islands. All islands sampled in our study are volcanic in origin so ostensibly contributions of naturally derived Hg should be similar across islands. In contrast, seawater in the far western Pacific has higher Hg levels compared to the Aleutians further to the east (Honda et al., 1990). While transport of Hg to the Aleutians via oceanic currents is complicated, seabirds in the western-most Aleutians may be exposed to elevated Hg particularly if they overwinter in the far western Pacific. Furthermore, Hg levels were 3–20 times higher in northern fulmars compared to glaucous-winged gulls and tufted puffins. This may reflect the long life span of northern fulmars (i.e. up to 40 years; Hatch and Nettleship, 1998) resulting in long term accumulation of demethylated Hg (Thompson and Furness, 1989; Elliot, 2005) magnified by its high trophic status. In addition, high concentrations of Hg in northern fulmars from our study were similar to those reported (≥10.0 μg/g d.w.) in other areas of the North Pacific (Honda et al., 1990; Elliot, 2005) as well as the Northwater Polynya (Campbell et al., 2005). Elevated concentrations of Hg in pelagic cormorants and pigeon guillemots likely reflect the piscivorous diets of these species.

4.2. PCB congener and chlorinated pesticide composition

The PCA on PCB congener concentrations revealed patterns not evident from examination of Σ PCBs alone and provided further evidence of a strong point source from Sweeper Cove at East Adak and a much weaker source from Constantine Harbor at Amchitka. More subtle yet important patterns in PCB congener composition were also indicated, whereby seabirds from the western islands of Buldir and Kiska typically had higher concentrations of lower chlorinated PCB congeners compared to seabirds from Adak. Perhaps the most striking pattern was the high Σ PCB concentrations detected in glaucous-winged gulls from Buldir and East Adak that clearly separated according to chlorination from low to high along Axis 2. Axis 2 explained only a small amount of the cumulative variance in PCB congener concentrations, yet a PCA analysis by Anthony et al. (2007) explained variation in PCB congeners in bald eagle eggs from the archipelago in a similar pattern and quantity, whereby eggs from Buldir and Kiska were characterized by lower chlorinated PCBs and eggs from East Adak by higher chlorinated PCBs. Seabird species likely vary in their capability to biotransform PCBs into persistent forms (Borga et al., 2005). Assuming biotransformation efficiencies within species do not change across islands, our results lend support for different sources of PCBs among islands. Lower chlorinated PCBs are more volatile and easily transported to higher latitudes via global distillation whereas highly chlorinated congeners especially PCBs 138 and 153 are more persistent and not easily metabolized (Van den Brink, 1997; Borga et al., 2001; Blais, 2005). These subtle differences in PCB congener composition may indicate that seabirds from Buldir and Kiska in the west were exposed to PCBs originating from a distant source, whereas high concentrations of more persistent higher chlorinated PCBs in seabirds from Adak serves as further evidence of a point source from East Adak. Conversely, differences in PCB congener composition may also be related to variable diets, particularly when scavenging seabirds such as northern fulmars, glaucous-winged gulls, and black-legged kittiwakes forage on carcasses of marine mammals resulting in increased exposure to persistent congeners (Wolkers et al., 1998; Borga et al., 2005).

Differences in DDE/Σ DDT ratios and composition of chlordane compounds provided further insight into contaminant sources in seabirds. For example, low DDE/Σ DDT ratios in tufted puffins from Buldir suggested some exposure to recently applied DDT from a distant source. Also, it was noteworthy that *p,p'* DDD (a parent DDT compound) comprised a high percentage of Σ DDT in trans-equatorial migrant short-tailed shearwaters and pelagically overwintering black-legged kittiwakes collected at Buldir. Overwintering movements to areas of the equatorial or far western Pacific waters may increase exposure to chlorinated pesticides. For example, non-resident seabirds and resident biota from Southeast Asia experience high exposure to DDT compounds (Minh et al., 2002).

Total chlordane in glaucous-winged gulls, tufted puffins, and most species from Buldir was comprised mostly of α (*cis*)-chlordane rather than γ (*trans*)-chlordane, which indicated exposure to weathered rather than recently applied chlordane (AMAP, 1998; Hung et al., 2002). Variability in chlordane composition is also related to phylogenic differences in the ability to biotransform technical chlordane into metabolic oxychlordane (Fisk et al., 2001). Accordingly, the metabolic by-product oxychlordane was the primary chlordane compound detected in northern fulmars, pigeon guillemots, and whiskered auklets, while larids and most alcids were not highly efficient at chlordane biotransformation (Fisk et al., 2001). Therefore, exposure to recently applied chlordane was unlikely for the species of seabirds in our study. While use of chlorinated pesticides has declined in the northern hemisphere, they are still in use in much of the world; especially in equatorial and developing countries (Hung et al., 2002; Koshy et al., 2006). Current and retrospective analyses of PCB, *p,p'* DDE, and Hg concentrations in black-footed (*Phoebastria nigripes*) and Laysan (*P. immutabilis*) albatrosses in the North Pacific suggested that overall contaminant levels may be increasing (Finkelstein et al., 2006). Our results indicate seabirds inhabiting the Aleutians are exposed mostly to environmentally persistent metabolites of chlorinated compounds, yet some exposure to recently applied DDT may occur for tufted puffins, black-legged kittiwake, and short-tailed shearwaters from Buldir.

4.3. Influence of trophic status and carbon source

The positive relationships between contaminant concentrations and $\delta^{15}N$ values we observed were expected given previously detected biomagnification of contaminants in high arctic (e.g. Atwell et al., 1998; Borga et al., 2001; Buckman et al., 2004) and lower latitude (Jarman et al., 1996) seabirds. Slopes obtained from regressing contaminant concentrations against $\delta^{15}N$ values are estimates of BMFs (e.g. Jarman et al., 1996; Borga et al., 2001; Buckman et al., 2004). Interestingly, BMFs for organic contaminants in our study were 2–3 times lower in comparison to a similar study of high arctic seabirds from Baffin Bay (Buckman et al., 2004), and the predictive power of these relationships in our study was also relatively low ($r^2 \leq 0.38$). Relationships between trophic status and contaminants often lack strong predictability, which may be attributed in part to the short-term metabolic turnover in skeletal muscle analyzed for stable isotopes compared to contaminants sequestered over long periods of time in liver tissue (Borga et al., 2005). Furthermore, the comparably weak BMFs observed in our study may reflect the complicated suite of factors affecting exposure to organic contaminants in Aleutian seabirds where the confounding effects of point and distant sources among islands and different migration patterns among seabirds lessens the impact of trophic status. Conversely, $\delta^{15}N$ may be a stronger predictor of contaminant burdens in high arctic seabirds because most contaminant exposure is derived from distant sources. If trophic status was the primary factor affecting accumulation of organic contaminants, then northern fulmars that had the most enriched $\delta^{15}N$ values would have the highest organic contaminant levels. This was not the case in our study. In contrast, BMFs for Hg in our study (0.26) were remarkably similar to others reported across numerous ecosystems that consistently range from 0.2–0.3 (Campbell et al., 2005). This similarity supports the hypothesis of Campbell et al. (2005) where consistent BMFs across ecosystems may be due to the close association between meHg and $\delta^{15}N$ in proteinaceous tissues, whereas OCs are associated with fatty tissues that are influenced more by local environmental variability.

The majority of contaminant and stable isotope studies have focused on trophic level effects, yet our results indicated that carbon sources can be important in explaining additional variation in contaminant concentrations among seabirds. While residual OC concentrations were not related to values of $\delta^{13}C$, an inverse relationship between residual Hg concentrations and values of $\delta^{13}C$ suggested higher Hg for seabird foraging in pelagic habitats. This provided further evidence for a distant source of Hg for some Aleutian seabirds. Inverse relations between Hg and $\delta^{13}C$ values have also been described for fish inhabiting high arctic lakes (Power et al., 2002). However, a larger sample of highly pelagic alcids and procellarids is necessary to more thoroughly explain relationships between pelagic production and Hg, as well as OCs.

Longitudinal enrichment of isotopic values owing to differences in baseline isotopic composition between islands (Schell et al., 1998) could bias relationships between contaminants and stable isotopes (Jardine et al., 2006). Although small sample sizes precluded using island as a covariate in our regressions, island differences in $\delta^{13}C$ values among glaucous-winged gulls and northern fulmars did not clearly change in a stepwise longitudinal fashion as expected based on large scale westward depletion of baseline $\delta^{13}C$ values across the archipelago (Schell et al., 1998). The lack of this pattern, which would imply foraging in local food webs with non-migratory prey (Rocque and Winker, 2004), further indicated dietary exposure to Hg of more distant origin.

5. Conclusion

Our results implicate 2 sources of contaminant exposure in Aleutian seabirds: local sources of contaminants associated with former military installations, and distant sources ostensibly owing to oceanic and atmospheric processes coupled with overwintering movements of seabirds that likely result in the transport of contaminants from industrialized regions of Eurasia and the far western Pacific. These conclusions are in general concordance with previous studies on organic contaminants and trace elements in biota from the archipelago (Anthony et al., 1999, 2007; Rocque and Winker, 2004).

Interesting patterns emerge when our results are qualitatively compared to reported concentrations in similar seabirds from different geographical regions, particularly high arctic latitudes, which aids in assessing possible health risks to seabirds and the organisms that consume them. For example, average Σ PCBs in glaucous-winged gulls from our study (1.1 μg/g) are at least twice as high as those reported in glaucous gulls (*Larus hyperboreus*), a similar and well studied congeneric species, from the Canadian high arctic (0.2 μg/g, Mallory et al., 2005) and Northwater Polynya (0.5 μg/g, Buckman et al., 2004), but still well below the high concentrations reported in glaucous gulls from highly contaminated regions of the Barents Sea (16.0 μg/g; Gabrielsen et al., 1995; 8.9–20.2 μg/g, Mehlum and Daealemans 1995). Similar patterns appear for Σ DDT in glaucous-winged gulls where concentrations in our study (0.2 μg/g) were similar to those reported in glaucous gulls from the Canadian arctic (0.1 μg/g, Mallory et al., 2005) and Northwater Polynya (0.4 μg/g, Buckman et al., 2004), but much lower than concentrations reported in glaucous gulls from other regions of the Barents Sea (3.5 μg/g, Gabrielsen et al., 1995). The high concentrations of Hg for northern fulmars in our study were similar to those reported in the North Pacific (15.9–16.7 μg/g, Honda et al., 1990; 10.5 μg/g, Elliot, 2005) and as a whole appear elevated in comparison to Hg concentrations in fulmars from the Barents Sea (2.0 μg/g, AMAP, 1998; 4.5 μg/g, AMAP, 1998). There may be risks to apex predators who consume seabirds, particularly procellariformes that are less sensitive to effects of high Hg (Thompson and Furness, 1989). Seabirds comprise a large percentage of bald eagle diets in the western Aleutians (Anthony et al., 1999, 2008). Given the high concentrations of Hg, as well OCs, detected in several seabirds from Buldir, it is intriguing that the small population of bald eagles on this island typically has depressed productivity (J. Williams and V Byrd, USFWS, pers. commun.). Additional work is needed to determine the mechanisms through which Aleutian seabirds are exposed to contaminants of distant origin (particularly Eurasia) and in turn may serve as a source of transport and exposure to local food webs (Blais et al., 2005).

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